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DETERMINATION OF SEVERAL AEROSOL DISTRIBUTIONS (U)

by

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ABSTRACT

A combination electrostatic classifier - nucleus counter was used to examine aerosols from three laboratory sources. A method of data reduction was devised, not requiring computational facilities, which appears adequate if the aerosol is not excessively heterogeneous.

RÉSUMÉ

Une combinaison de classificateur électrostatique - compteur à nucléation fut utilisée dans le but d'examiner des aérosols provenant de trois laboratoires. Une méthode de réduction des données a été développée ne nécessitant pas l'utilisation d'un ordinateur et celle-ci semble adéquate dans le cas où l'aérosol n'est pas excessivement hétérogène.

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INTRODUCTION

This technical note describes the use of two instruments, the electrostatic classifier (TSI 3071) and the condensation nucleus counter (CNC, TSI 3020), in the examination of aerosol size distributions. The classifier separates the aerosol into a series of narrow fractions on the basis of their electrical mobilities, and the CNC acts as a count instrument for the successive fractions.

The classifier-CNC combination was used to study aerosols generated by three sources:

(1) The TSI 3076 atomizer. This device, first described by Liu and Lee (1) is a development of the Collison atomizer with provision to ensure long term stability of output. Three of these are integral parts of the TSI FTS 400 system in use at DREO. For certain out-of-system applications, it was necessary to characterize the aerosol. In this work a solution of 0.2% w/w dioctyl phthalate (DOP) in isopropanol was used as source, the isopropanol evaporating from the aerial suspension to leave an aerosol of DOP droplets.

(2) The Q127 penetrometer (Air Techniques Incorporated, Baltimore, Maryland). This instrument is used primarily for canister testing, and is based on a technique first developed by Sinclair and La Mer (2). Liquid DOP is atomized and the original polydisperse spray is evaporated and recondensed to form an aerosol of considerably narrower size distribution. The output is claimed to be droplets of 0.3 µm average diameter, at a concentration of 100 ± 20 µg/l. This is either a mass median or mass mean diameter; which of these is not clear from the literature available on the instrument. In any case the system had been in continuous use with little maintenance for many years, and this work was a check to see whether its output still met specification requirements.

(3) The model 260 BC Respirator Fit Test System (Dynatech Frontier Corporation, Albuquerque, New Mexico). Here, liquid corn oil is atomized, and with no further treatment apart from removal of large droplets by an impactor, is taken up in an air stream and circulated through a test chamber. The size distribution then is expected to be broader than for the Q127. The mass mean diameter is stated to be in the range 0.5 - 0.7 µm, and the concentration with normal settings, 25 ± 5 µg/l.

It had been proposed to locate the Dynatech chamber in a cold room and operate it at temperatures ranging down to -20°C. The controls, aerosol generation and measurement systems would be of course in an adjoining room at ambient temperature, with air supply, return and sampling lines passing through ports into the cold room and thence to the test chamber itself. Comparative tests with the chamber at ambient conditions and under refrigeration were carried out to see the effect, if any, on the aerosol.

In addition to this, a brief examination was made of the laboratory air furnished by the building HVAC system. Results are only preliminary and semi-quantitative; one observes the expected dependence on the level of general activity in the building, in the submicron range, but there is an apparent increase in concentration as the size approaches 1 μm, which is unexpected and requires further investigation.

Several instruments have been described which measure aerosol size distributions (3-5). They all employ a mobility analyzer, such as the TSI 3071 classifier referred to above. The count instrument first employed was the electrical aerosol analyzer (6), but the CNC has more recently come into use.

When voltage and air flows have been established for the classifier, it will select, from the incoming aerosol, particles in a narrow range of electrical mobility. Unipositively charged particles in this mobility range have a correspondingly narrow range of particle diameter. Several stages of calculation, however, lie between the raw count data for successive voltage settings and the information ultimately sought, number concentration in each of the diameter intervals spanning the aerosol. The main complication is that aerosol particles exist in a number of charge states, so that with any given set of operating conditions, voltages and air flows, the classifier will select not merely singly charged particles of one size and the corresponding mobility, but also a fraction of doubly charged somewhat larger particles, which possess the same mobility; and likewise for triply charged, etc. Computer programs have been developed to treat the data by applying iteratively a series of corrections. The TSI company has marketed a system, the differential mobility particle sizer (DMPS, ref 7) consisting of the classifier and CNC, together with auxiliary equipment and a programme which executes a series of measurements, setting the required voltages and calculating distributions.

In general, the broader the dispersion the more extensive the data treatment needed to recover the size distribution. It was found here that a series of not very laborious hand calculations, to correct for doubly and triply charged particles, would give a quite reasonable approximation to the distribution, higher order corrections being negligible. This procedure was applied first to the Q127 and TSI 3076. Their outputs are reasonably homogeneous and centred around a size (several tenths of a micron) known to be well within the capacity of the classifier at reasonable air flows, so that the entire aerosol should be accessible to measurement. It was anticipated that the Dynatech output might not be equally amenable to treatment.

EXPERIMENTAL

AEROSOL SAMPLING

Aerosol was sampled from the Dynatech test chamber at atmospheric pressure. With the Q127, the hose conducting test aerosol to the canister chuck was detached, and connected to the sample (polydisperse) inlet of the classifier. The Q127 is an underpressure system, air being drawn by vacuum pump, and it appeared probable from the instrument manual that pressure at this point is near atmospheric. Sample flow, as will appear, is only a small fraction of the main (sheath air) flow, which is drawn directly from the ambient through a low pressure drop filter, so that the classifier is operating at atmospheric pressure in this case also.

The TSI atomizer was sampled through the large bore tygon tube which in normal operation of the FTS 400 conducts aerosol to the test chuck. This line passes three large capacity filters which introduce dilution air, and again will be nearly at atmospheric pressure.

For the room air sampling experiments a tygon tube 9.5 mm (3/8") i.d. by 1 metre long led from the classifier aerosol inlet and opened horizontally in the room at a height of seven feet. No further precautions, e.g. to secure isokinetic sampling, were felt necessary for this preliminary work.

ELECTROSTATIC CLASSIFIER

The theory of this instrument has been developed by Hoppel and others (8,9) and the expressions relating to its use as a mobility analyzer collected in convenient form in a TSI publication by Keady (7). The descriptions and derivations given in what follows are drawn entirely from these sources, and from the operating manual for the classifier. In brief, it has the form of a vertical cylindrical condenser, with inner and outer concentric stainless steel cylinders between which the aerosol passes. Clean (sheath) air is introduced at the top, and caused to flow laminarly down the space between outer cylinder and hollow central rod. The aerosol sample stream is introduced as a thin annular layer along the outer wall, avoiding turbulent mixing with the sheath air. Under the influence of an electric field applied between outer (+ve) and inner (-ve) cylinder,

positively charged particles are attracted toward the centre rod as the air streams downward. A narrow opening around the perimeter of the central rod, near its base, permits withdrawal of the fraction of aerosol of such mobility that, with voltage and air flows set, and the fixed dimensions of the apparatus, it just reaches the central rod at this point. The excess air is removed by a vacuum pump which maintains air flow in the system. Figure 1 shows the flows with the terminology used here. Q_a is evidently only a small fraction of the total air flow, Q_t. It is also apparent that the flows are connected by the equation:

$$Q_t = Q_c + Q_s = Q_e + Q_a \qquad [1]$$

Q_c, Q_e and Q_a are controlled by valves and these flows are measured by mass flowmeters. It is also necessary, in order to set air flows to their desired values, to install a valve on the sample inlet to control Q_s as well. To make calculations possible, the transfer function (to be defined below) must be symmetrical and triangular in form, as shown in Figure 2(a), and for this Q_a must be set equal to Q_s, from which it follows that $Q_c = Q_e$. As in fact the two latter flows are considerably larger than the former, the rough adjustment is made in reverse order, as follows.

With the clean air inlet opened wide, the vacuum pump is turned on and the excess air valve opened or closed until excess air flow is at the desired rate. Inlet (clean) air flow is now approximately the same as excess air flow. The CNC pump is turned on and the monodisperse aerosol valve is now opened to a flow of 0.25-0.30 l/min, approximately the intake of the CNC (0.3 l/min). The sample (polydisperse) valve is now opened to provide an adequate sample flow, and controls are then trimmed all round until $Q_e = Q_c$ (at the desired value, in the range 2.5 - 5 l/min) and accordingly $Q_a = Q_s$ at the considerably lower value ~0.25 - 0.30 l/min.

The larger the particle, the lower its electrical mobility, and the longer it will require to migrate from outer to inner cylinder. On the other hand, the lower the main flow rates Q_e and Q_c, the more time is available for this transport. Ideally these flow rates are set at values that will permit all the aerosol up to the largest particles to be detected as the voltage is varied from zero to its maximum value, near 11,000 volts.

It is clearly necessary that the charge distribution of the aerosol be known, so that for a given size, when the concentration in the +1 charge state is determined, the concentration of all particles in all charge states be calculable.

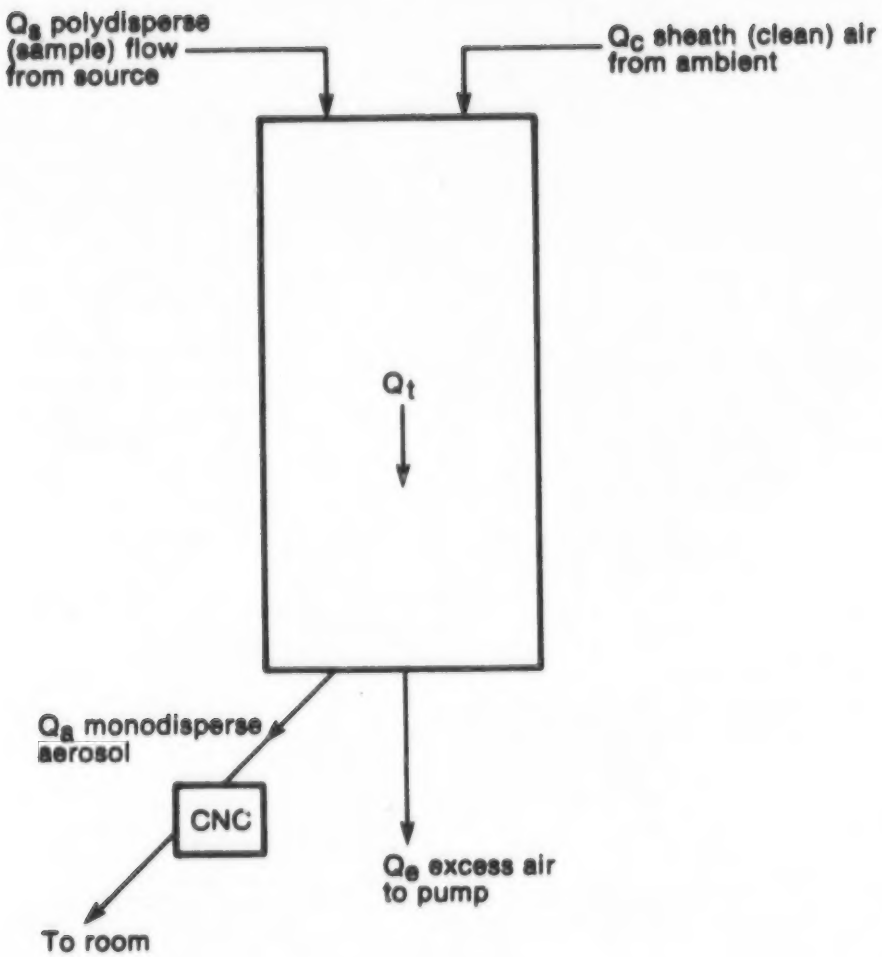
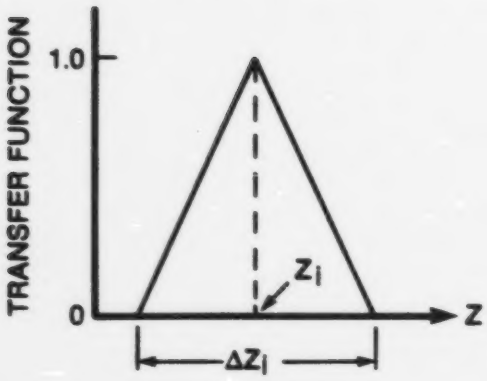
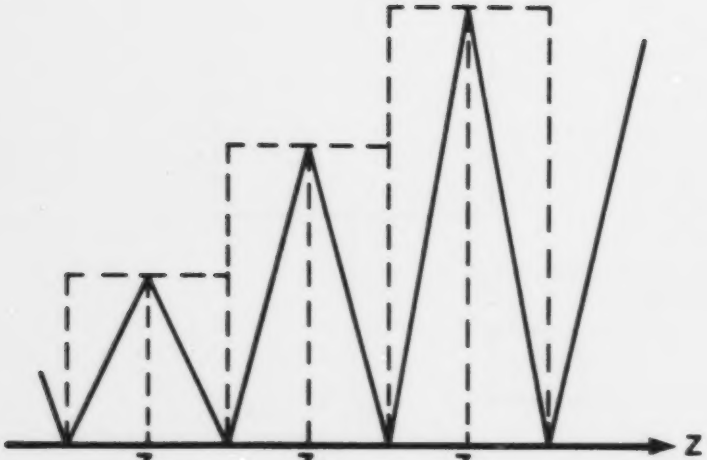


Figure 1: Schematic of electrostatic classifier, showing air flows.



(a)



(b)

Figure 2: (a) Transfer function for classifier under conditions described in text.

(b) Depiction of contiguous transfer functions, with proper choice of classifier voltage.

A Kr^{85} neutralizer in the inlet sample stream reduces the original random charge distribution of the aerosol to that corresponding to the Boltzmann equilibrium.

A recent calculation (10) of this distribution as a function of particle size appears in Table I. As expected, the distribution is symmetrical, with the uncharged component at each size always being the largest single fraction. By plotting these results as in Figure 3 one can interpolate to get the distribution at other diameters. It is to be noted that because of the polarity of the classifier, only positively charged particles are selected; also, that for a heterodisperse aerosol it is assumed that the particles in each size range adopt the distribution appropriate to that size, all these distributions coexisting in the aerosol.

TABLE I

Distribution of Charges on Aerosol Particles
According to Boltzmann's Law

D (μm)	Percent of particles carrying n elementary charge units								
	n_p = -4	-3	-2	-1	0	+1	+2	+3	+4
0.01				0.34	99.32	0.34			
0.02				5.23	89.53	5.23			
0.04			0.23	16.22	67.10	16.22	0.23		
0.06		0.01	1.25	21.30	54.88	21.30	1.25	0.01	
0.08		0.08	2.78	23.37	47.53	23.37	2.78	0.08	
0.10		0.26	4.39	24.09	42.52	24.09	4.39	0.26	
0.20	0.32	2.33	9.66	22.63	30.06	22.63	9.66	2.33	0.32
0.40	2.19	5.92	12.05	18.44	21.26	18.44	12.05	5.92	2.19
0.60	3.82	7.41	11.89	15.79	17.36	15.79	11.89	7.41	3.82
0.80	4.83	7.94	11.32	14.00	15.03	14.00	11.32	7.94	4.83
1.00	5.42	8.06	10.71	12.70	13.45	12.70	10.71	8.06	5.42

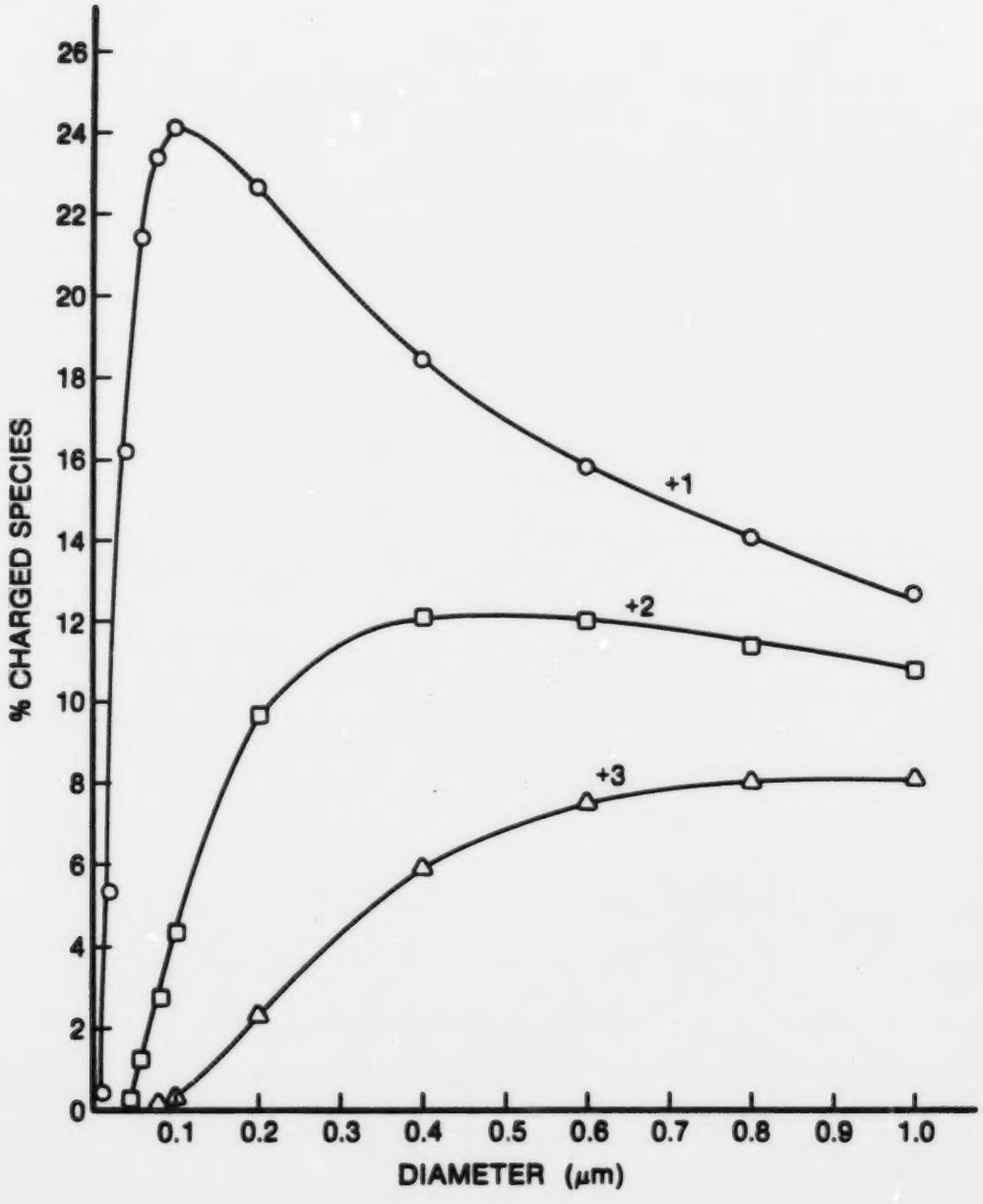


Figure 3: Boltzmann equilibrium distribution of charge in aerosols, as a function of size.

In the DMPS arrangement described by TSI, the CNC outlet leads back into the Q_e flow line through a regulating valve, so that the CNC is part of a closed loop. However, for the CNC to operate properly, air flow through it, monitored by an analog meter, must be maintained accurately at 5 ml/s or 0.3 l/min. With the TSI arrangement it was found impossible to work out any valve settings in which the much larger flow Q_e did not over-ride the relatively small capacity pump that moves air through the CNC. Accordingly the arrangement of Figure 1, with the CNC exhausting to the atmosphere, was used.

THEORY OF CLASSIFIER OPERATION

The electrical mobility Z of a particle in the classifier field is expressed in units of cm/sec per volt/cm or velocity per unit field strength. It has been shown (ref 7) that the mobility selected by the classifier under a given set of operating conditions is related to the other parameters by,

$$Z = \frac{[Q_t - \tfrac{1}{2}(Q_s + Q_a)] \ln \frac{r_2}{r_1}}{2\pi VL} \qquad [2]$$

in which the flows are (in this derivation) expressed in cm^3/sec, r_1 and r_2 are the inner and outer radii of the classifier, V the voltage and L the passage length from sample inlet to monodisperse aerosol outlet. Under given conditions, ZV=K, and as V is increased particles of successively smaller mobility (i.e. larger size) are selected. Mobility, or velocity/unit force is expressible in another way from the Stokes equation $F = \frac{3\pi\eta D v}{C}$, the symbols having the usual significance, (note that small v here is velocity, to be distinguished from voltage V). C is the Cunningham correction which becomes important in the submicron region. $\frac{v}{F}$ varies inversely with diameter; from these equations a relation between voltage and size selected is at once derivable.

The selected mobility has a finite width

$$\Delta Z = \frac{(Q_s + Q_a) \ln \frac{r_2}{r_1}}{2\pi VL} \qquad [3]$$

Within this width the probability of selection of a particle is expressed by a transfer function which varies from 1 at Z, the central value to zero at $Z \pm \frac{\Delta Z}{2}$, the extremes. It is thus representable as a triangular area (Figure 2(a)). It is also seen that only half the particles in the mobility band ΔZ are selected. If this analysis is correct, then by choosing voltages properly one can divide up the aerosol into a series of contiguous mobility bands, and the total measured particle count will be half the actual concentration of the aerosol.

The condition for contiguous mobility bands is, referring to Figure 2(b), that,

$$Z_2 - Z_1 = \tfrac{1}{2} (\Delta Z_2 + \Delta Z_1) \qquad [4]$$

Referring to equations 2 and 3,

$$Z = \frac{Ka}{V} \quad \text{where } Ka = \frac{[Q_t - \tfrac{1}{2} (Q_s + Q_a)] \ln \frac{r_2}{r_1}}{2\pi L}$$

hence $$Z_1 = \frac{Ka}{V_1}, \quad Z_2 = \frac{Ka}{V_2}$$

and $$\Delta Z = \frac{Kb}{V} \quad \text{where } Kb = \frac{(Q_s + Q_a) \ln \frac{r_2}{r_1}}{2\pi L}$$

and $$\Delta Z_1 = \frac{Kb}{V_1}, \quad \Delta Z_2 = \frac{Kb}{V_2}$$

Substituting in equation (4), one finds

$$\frac{V_2}{V_1} = \frac{Ka - Kb/2}{Ka + Kb/2} \quad \text{which reduces to}$$

$$\frac{V_2}{V_1} = 1 - \frac{Q_s + Q_a}{Q_t} \qquad [5]$$

a dimensionless ratio. Hence for any set of flows defined by Q_s, Q_a and Q_t, a series of voltages in geometric progression will survey the entire aerosol in mobility bands touching at the base as in Figure 2(b).

GENERAL PROCEDURE

Operation of the classifier and CNC is described in company manuals. The system shown schematically in Figure 1 was set up, using a Gast pump to draw air. When measuring instruments and air flows had stabilized, the classifier was set at a series of voltages and the resulting CNC readings noted. This entailed recording by hand and averaging readings which fluctuated about a final steady value. The procedure for converting raw data into finished results is rather complex; accordingly one example is worked out in nearly complete detail in Appendix A. In this example, the TSI 3076 atomizer was used as source, with $Q_c = Q_e = 3$ l/min, and $Q_a = Q_s = 0.25$ l/min, so that $Q_t = 3.25$ l/min.

From equation 5 it is seen that to cover the entire aerosol with contiguous mobility bands the ratio of successive voltages $R = 1 - \frac{Q_s + Q_a}{Q_t}$, which in this case equals 0.8461. Thus if the final highest voltage is set at 10000V, the sequence will be 10000, 10000R, 10000R^2 etc. or 10000, 8401, 7159, 6057 etc. volts, as shown in Table A-1. Usually measurements were taken with increasing voltages, starting at a value at which count readings first became significant. Repeat runs and prolonged runs at a single voltage were made at intervals, as a check on system stability.

The accuracy of sizing was investigated, using highly monodisperse aerosols of polystyrene latex spheres (PSL). These were nebulized in the FTS 400 and after drying passed into the classifier, the voltage being adjusted until the CNC count reached a maximum. From equation 2 the mobility Z was calculated, and hence the particle diameter, as explained in Appendix A. Measured diameters at four flow rates are listed below.

TABLE II

Flow Rates	Measured Diameter			
2.5 l/min	1.00	.59	.42	.32
3.0 l/min	-	.58	.42	.32
4.0 l/min	-	.57	.41	.31
5.0 l/min	-	-	.41	.31
Nominal diameter, μm	.916	.562	.399	.305

Because of the lower size cutoff limits at higher flow rates, the larger sizes become inaccessible to measurement. Agreement, apart from the .916 mμ diameter at 2.5 l/min, appears to be satisfactory. The concentration maxima observed are not as sharp as might be expected from the standard deviations on latex diameter quoted by the manufacturer; however these numbers are obtained by electron microscopy, while the PSL employed here may have undergone some swelling, and is not completely dried out during aerosol preparation.

DYNATECH OPERATION

The 3076 atomizer and Q127 were sampled as described earlier. With the Dynatech, sampling was done through a 9.5 mm (3/8") i.d. tygon tube, about 3 metres in length, leading from the centre of the chamber through the cold room to the classifier, which was placed immediately next to the cold room to minimize air paths.

In the operation of this test system air is circulated in a closed cycle between the control module and the test chamber, the aerosol being injected into the main airstream in the test chamber inlet line. The emergent air stream is filtered to remove aerosol prior to regeneration, a part of the return air being diverted through the atomizer and then rejoining the main stream. With the cold room at -20°C, the return air had no opportunity to warm up, and it was felt that this might affect operation of the atomizer, perhaps by cooling the corn oil reservoir and affecting its viscosity. Accordingly runs at -20°C were conducted, first as described above, and then with the return air line in a vessel of lukewarm water.

TEST CONDITIONS

The following runs were conducted:

(i) Q127 3, 4, 5, l/min

(ii) TSI 3076 3, 5 l/min

(iii) Dynatech RT (-19°C) 2.5 3 4 l/min
0°C 3 l/min
-20°C 3 l/min - no reheat of return air
3 l/min - with reheat of return air

(iv) Room air 2.5 l/min 1530-1630 Weekday
2030-2130 Saturday
0830-0930 Weekday

RESULTS AND DISCUSSION

In Appendix A the experimental results for TSI 3076 at a classifier airflow of 3 l/min were worked out in detail and shown in Figure 4. Results at 5 l/min are similarly shown in Figure 5 and calculations for all the 3076 atomizer work summarized in Table III(a). Comparing the figures, it is seen that the lower flow rate, with larger upper cut-off size, covers the aerosol more completely. This is reflected in the slightly larger values for mass mean and median diameters. Second and third order corrections are also seen to be smaller at 3 l/min (see Figure 4).

This is presumably because at the lower flow rate the upper size cutoff is larger (.840 μm cf .548 μm) and so the assumptions made in the calculations reflect more closely the actual situation. From Figures 4 and 5 it is seen that the ratio $\frac{\text{concentration at largest size}}{\text{peak concentration}}$ is, in the first approximation, about 5% at 5 l/min, but less than 0.5% at 3 l/min.

Results for the Q127 are summarized in Table III(b). Mean and median diameters all lie in the range 0.32 - 0.39 μm, somewhat larger than the 0.3 μm stipulated. As with the TSI 3076, which also has a reasonably monodisperse aerosol, the first approximation does not differ much from the final corrected distribution at 3 l/min. Results are plotted in Figure 6. It was not felt worthwhile to make higher order corrections for the two larger flow rates, 4 and 5 l/min.

Data for the Dynatech appear in Table III(c), and the more interesting results are plotted in Figures 7 and 8. Figure 7 shows the first order approximation aerosol distribution at three temperatures, the experiment at -20°C being conducted both with and without reheat of the return air. Qualitatively the shapes are similar, and the quantitative count difference is evidently an artifact of the return air temperature. This similarity is confirmed by examination of mean and median diameters listed in Table III(c); in particular it is seen on comparing results for 3 l/min at 20°C and at -20°C with reheat. Plots of concentration (1st approximation and 3rd order correction) against size for these two runs are nearly superimposable.

TABLE III

Summary of derived aerosol properties[1]

In the first column, A signifies first order approximation,
B - third order correction

Order	Maximum Size Measured μm	Maximum (Mode) in Number Dist μm	Number Conc At Max	Number Mean Diameter μm	Mass Median Diameter μm	Mass Mean Diameter μm	Calculated Conc μg/l
			(a) TSI 3076 Atomizer, 3 l/min				
A	.840	.135-.150	2.50+6	.16	.29	.34	125
B	.840	.167	1.94+6	.18	.30	.36	110
			TSI 3076 Atomizer, 5 l/min				
A	.548	.16	7.91+5	.18	.29	.31	80
B	.548	.16-.18	5.20+5	.20	.31	.33	70
			(b) Q127 3 l/min				
A	.840	.17	2.76+6	.21	.32	.37	200
B	.840	.21	2.26+6	.23	.34	.39	170
			Q127 4 l/min				
A	.686	.20	9.20+5	.22	.34	.38	125
			Q125 5 l/min				
A	.548	.22-.24	2.78+5	.24	.34	.36	50
			(c) Dynatech 3 l/min R.T.				
A	.840	.26	9.48+4	.28	.51	.54	29
B	.840	.37	6.03+4	.34	.55	.58	25
			2.5 l/min R.T.				
A	.983	.28	9.05+4	.29	.54-.55	.59	26
B	.983	.32	5.44+4	.33	.59	.63	22
			4 l/min R.T.				
A	.686	.29	6.58+4	.29	.46	.47	21
			3 l/min 0°C				
A	.840	.26	6.27+4	.29	.51	.53	19
			3 l/min -20°C No Reheat				
A	.84	.26	5.39+4	.29	.50	.54	13
			3 l/min -20°C Plus Reheat				
A	.840	.29	8.74+4	.31	.52	.55	31
B	.840	.37	5.87+4	.38	.55	.59	27

Note 1: See footnote, Table A-1 (Appendix A), for method of expressing number concentrations as powers of 10.

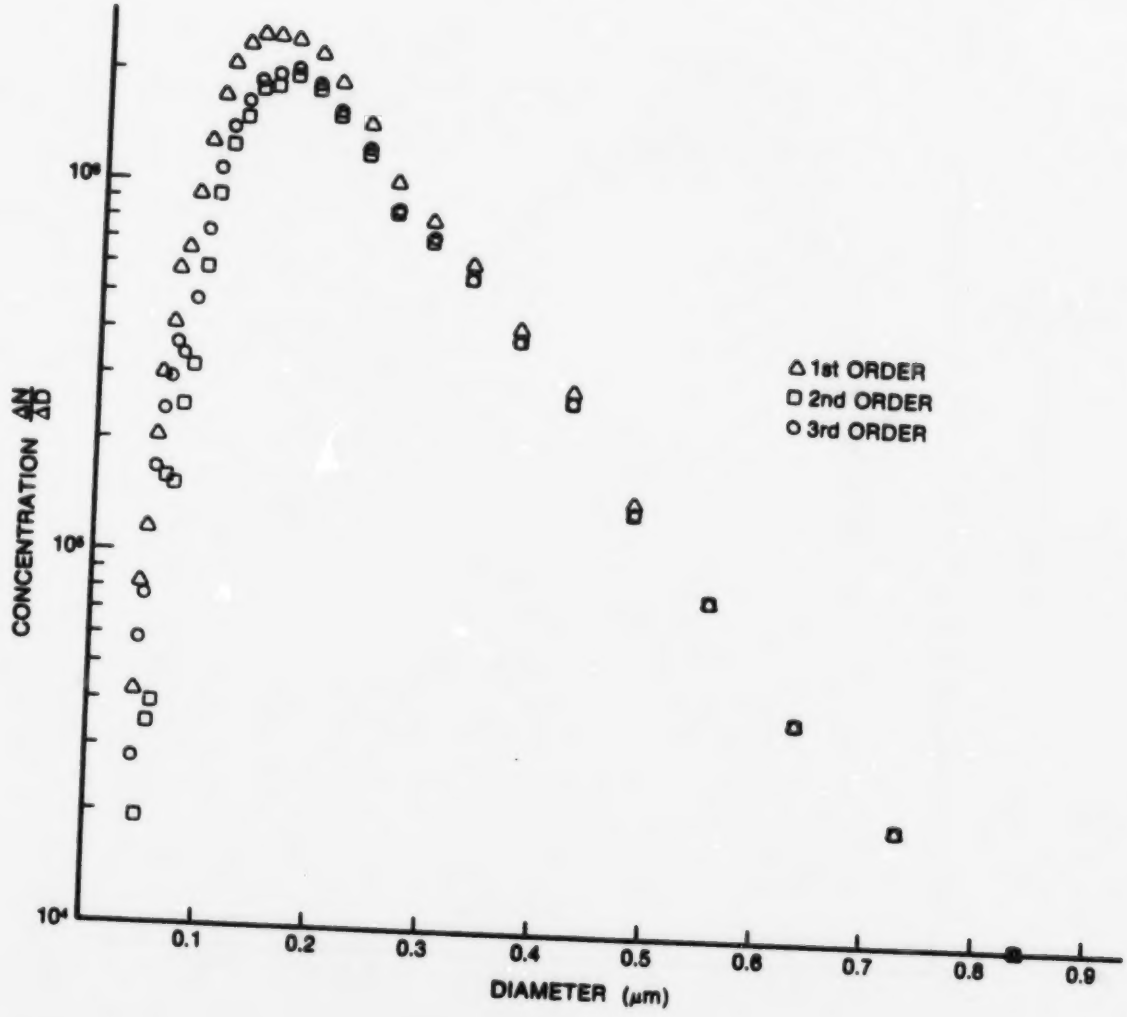


Figure 4: Size distribution, TSI 3076 atomizer, 3 l/min, several orders of approximation.

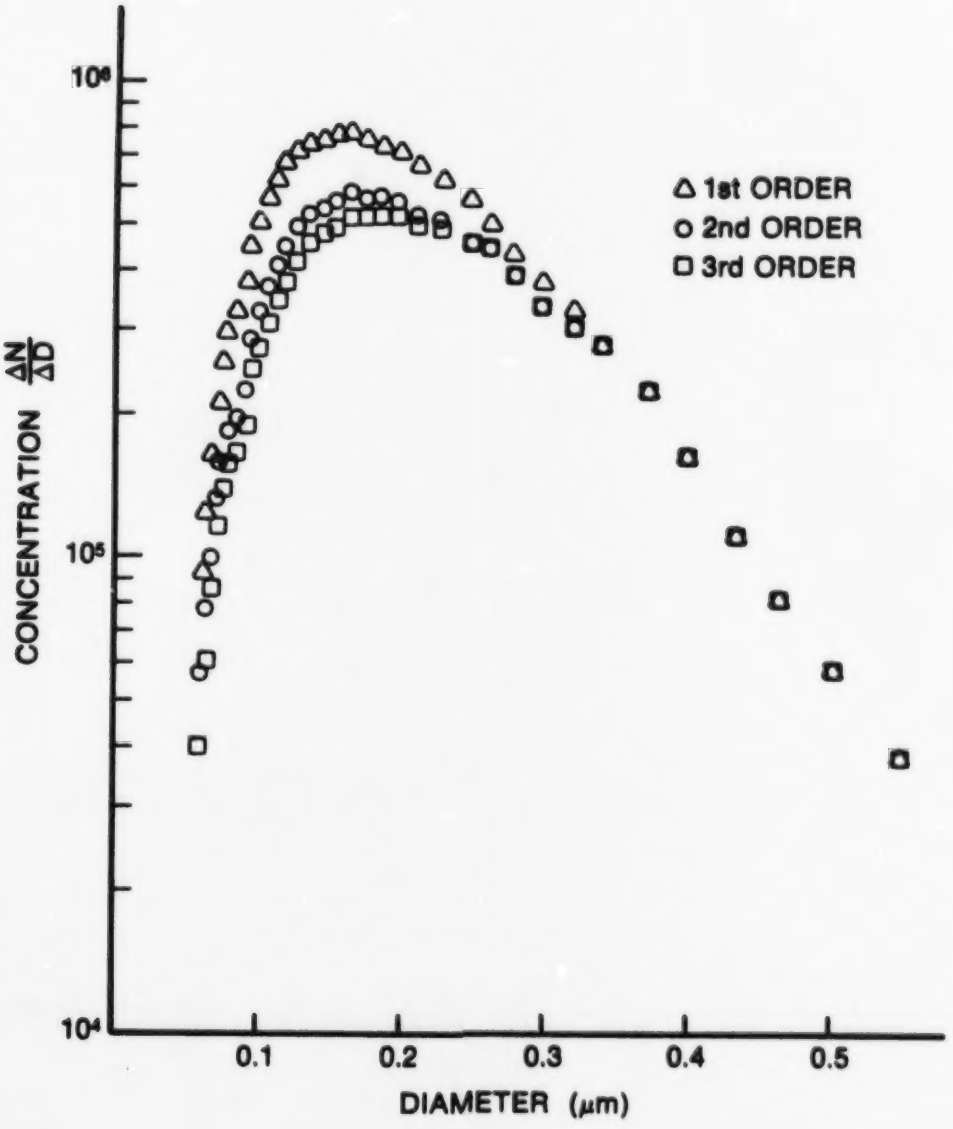


Figure 5: Size distribution, TSI 3076 atomizer, 5 l/min, several orders of approximation.

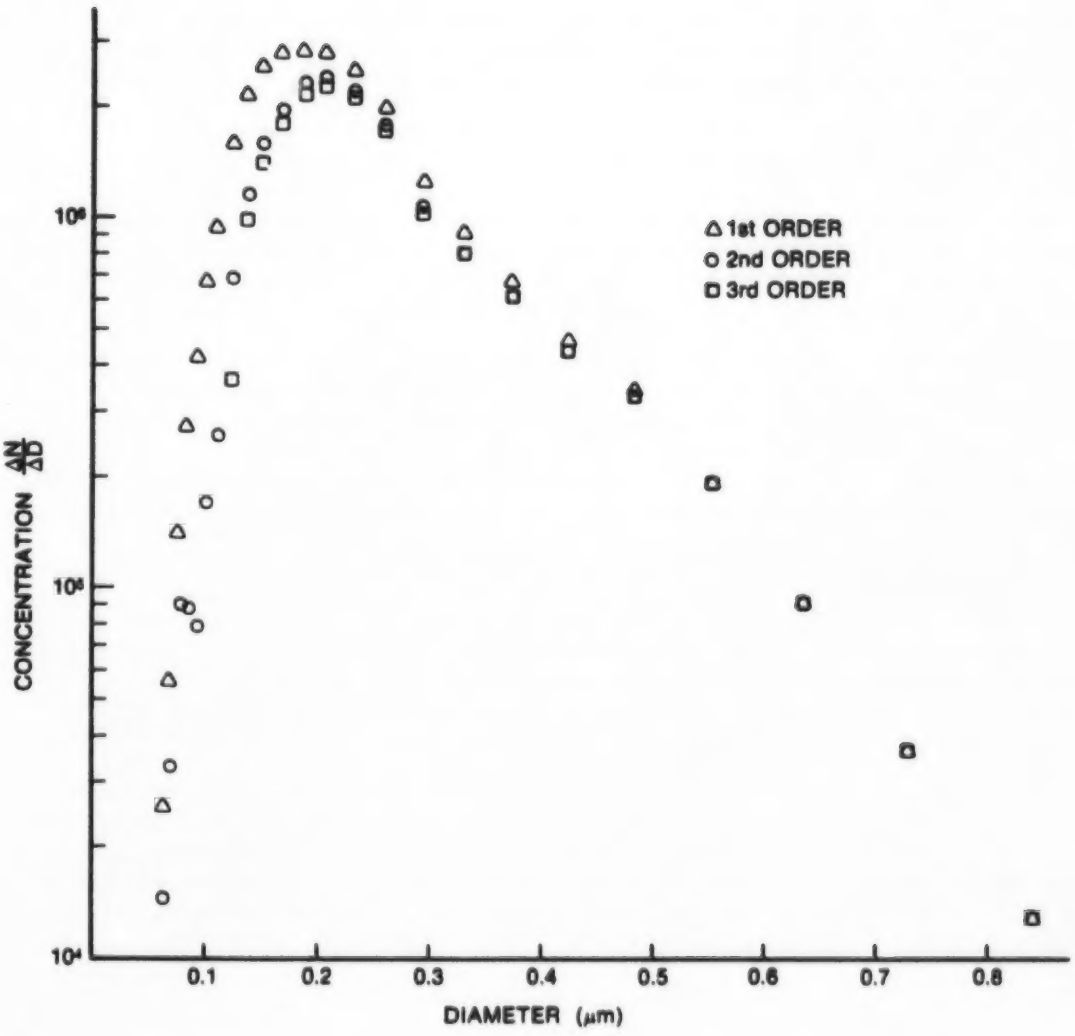


Figure 6: Size distribution, Q127, 3 l/min, several orders of approximation.

The stated mass mean diameter for the Dynatech is 0.5 - 0.7 μm. All values listed in Table III(c) lie in the range 0.50 - 0.63. This diameter is considerably greater than the two previous ones, and even at the lowest practical flow rate, 2.5 l/min, a considerable fraction is cut off at the upper end (see Figure 8). This is reflected in a sizable difference in calculated mean and median diameters on passing from 2.5 to 3.0 l/min and the effect is even more noticeable at 4 l/min. At room temperature, and also at -20°C when the return air is reheated, the calculated mass concentrations all lie in the range 25 ± 5 μg/l, as stipulated.

Results of the three ambient air sampling experiments are plotted on Figure 9, as first order approximations (against log diameter in this case, because of the spread in size). The appearance of a fraction apparently peaking somewhere above 1 μm requires further study.

The reliability of the mass concentrations in the last column of Table III is questionable. In principle this measurement should be independent of flow rate, so long as this latter is not so great that an appreciable fraction of aerosol at the upper size end is missed. Results for the Dynatech, when cooling problems were absent, were as expected, and were independent of flow; however for the TSI 3076 and the Q127, results were sensitive to flow rates. With the Q127, calculated concentrations varied from ~ 200 μg/l to ~ 50 μg/l on passing from 3 l/min to 5 l/min, as compared with the stipulated 100 ± 20 μg/l. A similar but smaller variation was observed with the TSI 3076. Using this latter source, a direct determination of aerosol concentration was carried out, passing the aerosol stream at known flow rates through a filter and measuring the increase in weight. At 3 and 5 l/min the concentrations found by weight increase were 80-85 and 52-57 μg/l respectively, as compared with 110 and 70 μg/l reported in Table III. Perhaps in view of the highly indirect method of measurement, effectively integrating over a series of diameter intervals, agreement within a factor of two is all that can be expected. It is of interest that summing the particle counts in the atmospheric experiments leads to a value slightly larger than $10^4/cm^3$, about 50-60% of the count obtained when the CNC sampled the laboratory air directly. Figure 9 indicates that a sizable fraction ≥ 1 μm is being missed.

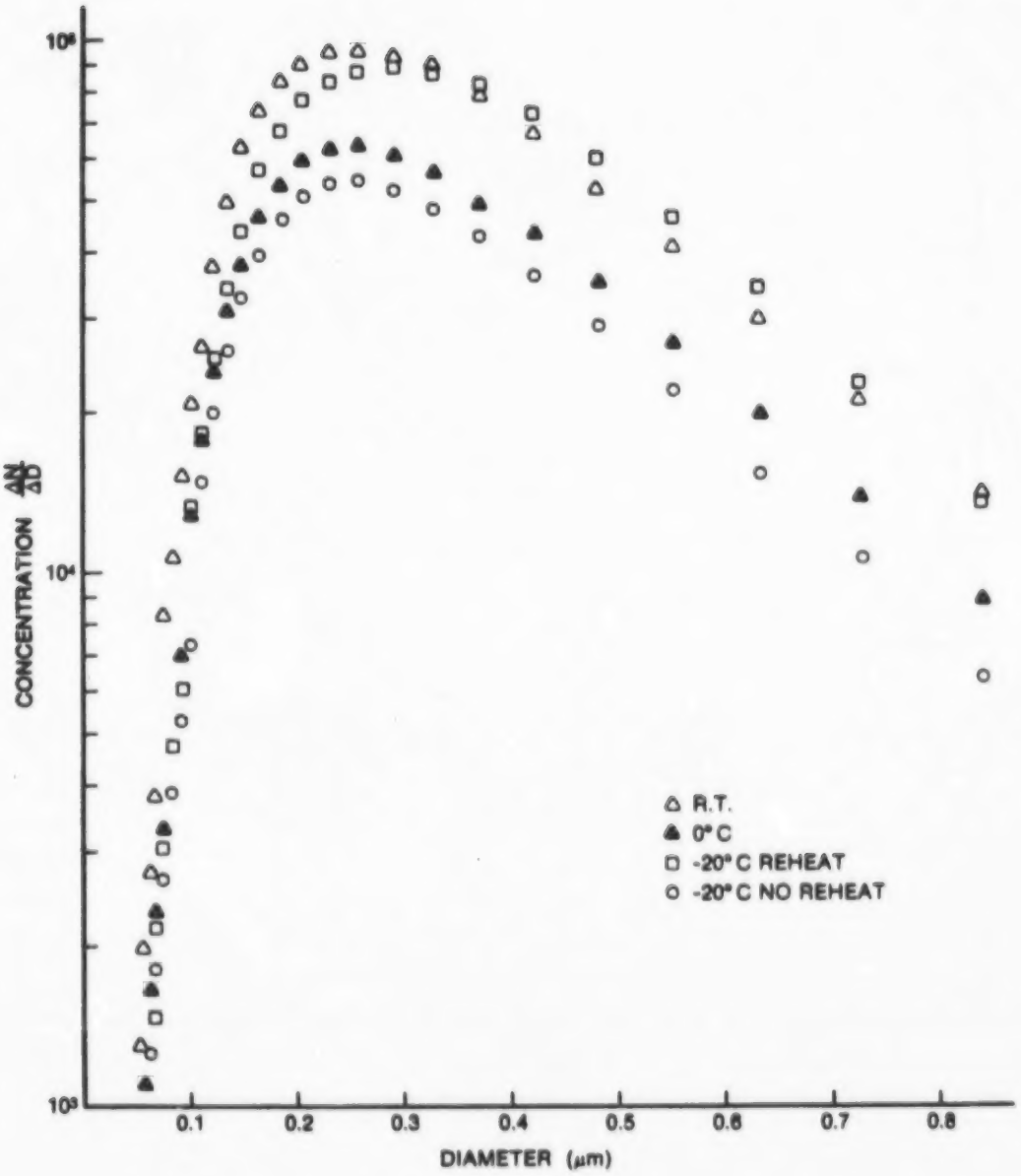


Figure 7: Size distribution, Dynatech, 3 l/min, at several temperatures.

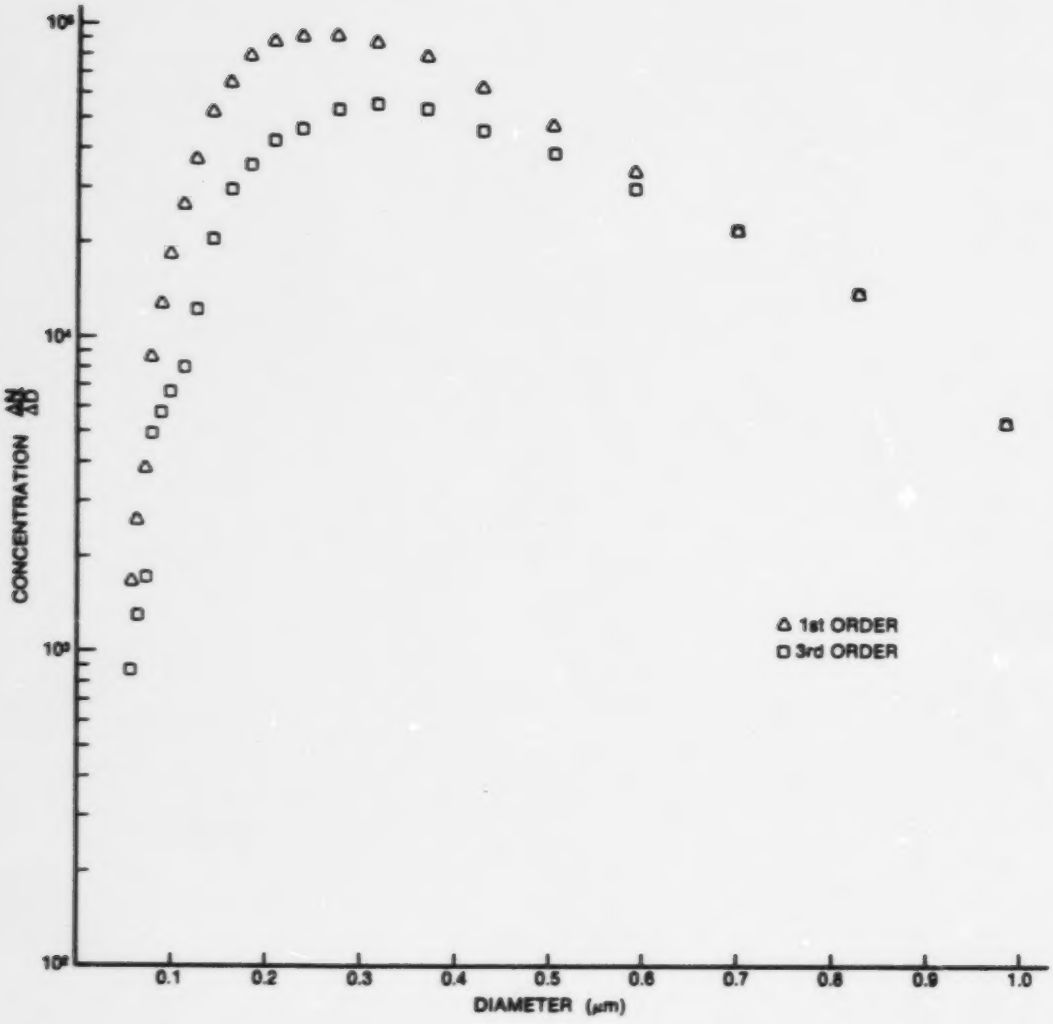


Figure 8: Size distribution, Dynatech, 2.5 l/min, two orders of approximation.

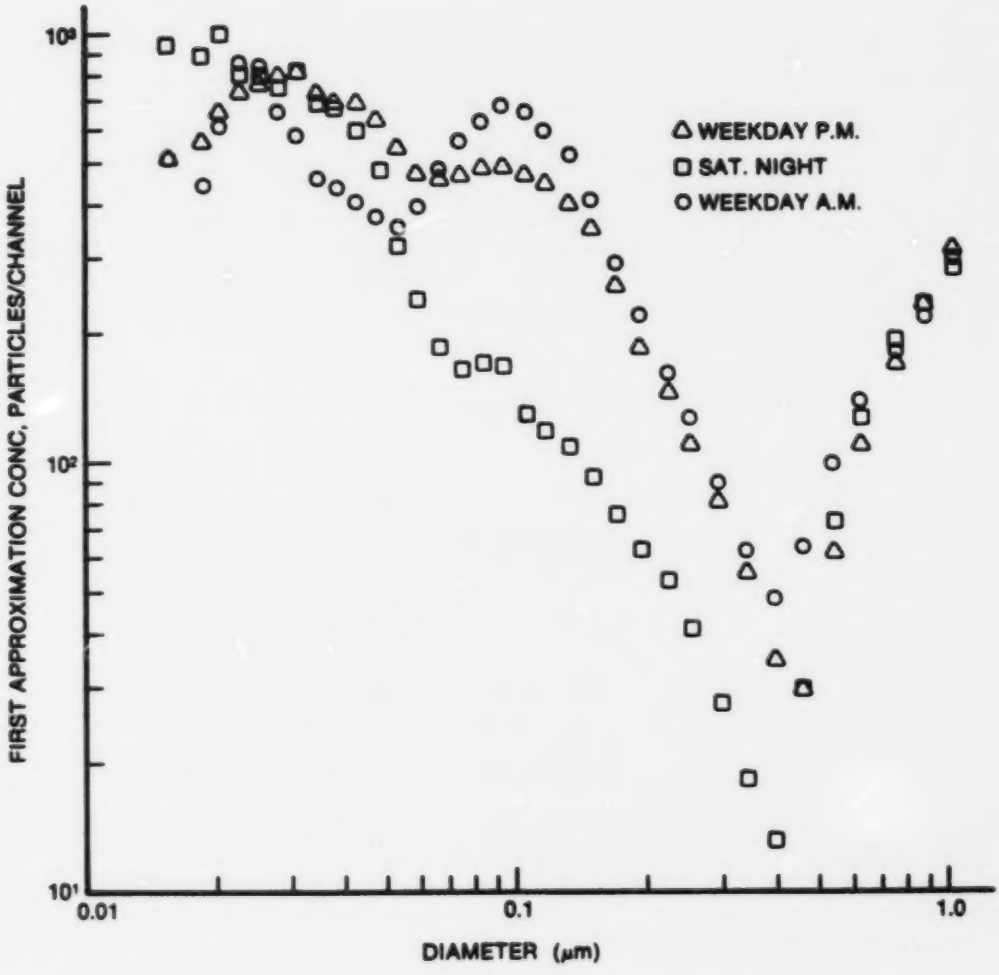


Figure 9: Atmospheric aerosol (laboratory), preliminary results, taken at several times.

SUMMARY AND CONCLUSION

Use of the electrostatic classifier and nucleus counter in tandem, a well-established technique for determining aerosol size distributions, provided information on the output of three laboratory generators that agreed reasonably well with manufacturer's claims. Mean diameters for the Q127 and Dynatech aerosols were close to the values specified. Weight concentrations were at worst within a factor of two of the expected values. A preliminary examination was made of an uncontrolled laboratory atmosphere. Caution must be exercised in interpreting results with broadly dispersed aerosols, as from the Dynatech. The method described here for determining higher order corrections is convenient for occasional or one-time use.

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APPENDIX A

DETAILS OF CALCULATION

GENERAL

It is desirable that the entire aerosol size range be spanned, so that at the highest classifier voltage (corresponding to the lowest mobility range and the largest diameter) the CNC count be nearly zero, or at least much smaller than the peak concentration. Then one is assured that there is only a negligible number of still larger particles of +2 or higher charge falling in these channels; i.e. it can be assumed that the entire contents of these channels are +1 charged particles of the appropriate size. This condition is approximated in the TSI 3076 and the Q127 atomizers, and facilitates calculation. As seen above, reducing the air flow rate enables one to push up the upper size cutoff and record larger particles. A practical limitation is the voltage-flow characteristics of the mass flowmeters; these become increasingly sensitive at low flow rates; one cannot go below a rate of $Q_c = Q_e = 2.5$ l/min, corresponding to a maximum diameter of ~ 1.0 μm. At this flow rate the excess air valve is barely cracked open.

DETAILED CALCULATION FOR THE TSI 3076 ATOMIZER, $Q_c = Q_e = 3$ l/min

First Order Approximation

This assumes that only +1 charged particles are present throughout, and from the raw CNC count at each voltage corresponding to a mobility and therefore to a particle size, uses the Boltzmann equilibrium distribution to calculate the total concentration at this size. In the following table (A-1), each column has seven entries.

(1) Voltage

(2) Raw CNC count

(3) Mobility Z corresponding to voltage (1), calculated from equation 2 above; here, for $Q_c = Q_e = 3$ l/min, and $Q_s = Q_a = 0.25$ l/min, $Z = 0.1319/V$.

(4) Particle size (μm) corresponding to mobility. Values of Z vs D have been calculated; a table covering the range D = 0.001 to 1.0 μm appears in the manual for the TSI 3071.

(5) The raw count in (2) is doubled, in order to calculate the total number count. As pointed out above, the triangular form of the transfer function results in only half the total aerosol in each mobility channel being counted. In subsequent calculations of aerosol distribution, mass mean diameter, etc., the number count per channel derived from (5) appears twice in compensating ways, and so these average values are unaffected by doubling all counts.

(6) The % +1 charge at Boltzmann equilibrium for the size in (4) is determined by interpolating in Figure 3.

(7) The raw count (5) is divided by (6) to give a first approximation to aerosol concentration at size (4). More correctly the diameter in (4) is a diameter range corresponding to the mobility range ΔZ, and centred on the nominal value given. All plots in the graphs which follow, whether first approximations or higher order corrections, are concentrations at a series of diameter ranges.

Results for the TSI 3076 are listed in the second column of Table A-2 and plotted as the upper curve in Figure 4. It is apparent that to this approximation the concentration at the largest size measured (>0.8 μm) is a small fraction of the maximum, at 0.13 - 0.15 μm.

Second Order Calculation

The mobility of a doubly charged particle is twice that of a singly charged particle of the same size. As mentioned above, it is assumed of necessity that the upper channels (diameters) are populated only by +1 charged particles of the stated diameter.

Inspecting Table A-1, it is seen that the mobility of .840 μm particles (1.319 - 5) on doubling to (2.638 - 5) lies between the values for .483 μm (2.574 - 5) and .424 μm (3.042 - 5), and closer to the former. Then +2 charged particles of .840 μm diameter will be disposed primarily in these two channels, and mostly at .483 μm. In the approximation used here, all the contribution of +2 charged particles of 0.840 mμ diameter is attributed to this smaller size. Obviously in computer calculations this contribution would be spread over several channels, taking account of the fact that mobility widths are a function of diameter.

TABLE A-1

TSI 3076, 3 l/min. Derivation of first order approximation, concentration as a function of size.

1.	93	110	130	153	181
2.	3.4 + 3	7.00 + 3	1.03 + 4	2.05 + 4	3.12 + 4
3.	1.418 - 3[a]	1.199 - 3	1.014 - 3	8.62 - 4	7.29 - 4
4.	.040	.0438	.048	.0523	.0572
5.	6.8 + 3	1.40 + 4	2.06 + 4	4.10 + 4	6.24 + 4
6.	16.2	17	18	20.2	20.8
7.	4.2 + 4[a]	8.2 + 4	1.14 + 5	2.03 + 5	3.00 + 5

1.	214	253	299	354	418
2.	4.38 + 4	6.35 + 4	8.46 + 4	1.07 + 5	1.52 + 5
3.	6.16 - 4	5.21 - 4	4.41 - 4	3.72 - 4	3.16 - 4
4.	.0627	.0688	.0755	.0831	.0912
5.	8.76 + 4	1.27 + 5	1.492 + 5	2.14 + 5	3.04 + 5
6.	21.5	22.4	23.0	23.5	23.8
7.	4.08 + 5	5.67 + 5	6.49 + 5	9.10 + 5	1.28 + 6

1.	494	584	690	815	964
2.	2.05 + 5	2.52 + 5	2.82 + 5	2.98 + 5	2.94 + 5
3.	2.64 - 4	2.26 - 4	1.91 - 4	1.618 - 4	1.37 - 4
4.	.101	.111	.123	.136	.150
5.	4.10 + 5	5.04 + 5	5.64 + 5	5.96 + 5	5.88 + 5
6.	24.1	24.1	24.0	23.8	23.5
7.	1.70 + 6	2.09 + 6	2.35 + 6	2.50 + 6	2.50 + 6

1.	1139	1346	1591	1880	2222
2.	2.82 + 5	2.54 + 5	2.10 + 5	1.60 + 5	1.08 + 5
3.	1.158 - 4	9.80 - 5	8.29 - 5	7.016 - 5	5.936 - 5
4.	.167	.187	.207	.232	.260
5.	5.64 + 5	5.08 + 5	4.20 + 5	3.20 + 5	2.16 + 5
6.	23.2	22.9	22.4	21.9	21.3
7.	2.43 + 6	2.22 + 6	1.88 + 6	1.46 + 6	1.01 + 6

1.	2626	3104	3669	4336	5125
2.	8.32 + 4	6.09 + 4	3.92 + 4	2.55 + 4	1.24 + 4
3.	5.023 - 5	4.249 - 5	3.595 - 5	3.042 - 5	2.574 - 5
4.	.293	.330	.373	.424	.483
5.	1.664 + 5	1.218 + 5	7.84 + 4	5.10 + 4	2.48 + 4
6.	20.7	19.8	18.9	18.0	17.2
7.	8.04 + 5	6.15 + 5	4.15 + 5	2.83 + 5	1.44 + 5

TABLE A-1 (Cont'd)

1.	6057	7159	8461	10000
2.	6.55 + 3	2.98 + 3	1.50 + 3	6.8 + 2
3.	2.178 - 5	1.842 - 5	1.559 - 5	1.319 - 5
4.	.552	.633	.728	.840
5.	1.31 + 4	5.96 + 3	3.0 + 3	1.36 + 3
6.	16.3	15.4	14.6	13.7
7.	8.04 + 4	3.87 + 4	2.05 + 4	1.0 + 4

(a) In what follows, to simplify the tabular presentations, positive and negative exponents of ten are indicated as shown. Thus 1.418 - 3 = 1.418×10^{-3}, 4.2 + 4 = 4.2×10^{4}.

TABLE A-2

TSI 3076, 3 l/min. Concentration as a function of size.

Size, μm	1st Order	2nd Order	3rd Order
.840	1.0 + 4	1.0 + 4	1.0 + 4
.728	2.05 + 4	2.05 + 4	2.05 + 4
.633	3.87 + 4	3.87 + 4	3.87 + 4
.552	8.04 + 4	8.04 + 4	8.04 + 4
.483	1.44 + 5	1.38 + 5	1.38 + 5
.424	2.83 + 5	2.70 + 5	2.70 + 5
.373	4.15 + 5	3.90 + 5	3.90 + 5
.330	6.15 + 5	5.66 + 5	5.66 + 5
.293	8.04 + 5	7.25 + 5	7.15 + 5
.260	1.01 + 6	8.59 + 5	8.45 + 5
.232	1.46 + 6	1.25 + 6	1.22 + 6
.207	1.88 + 6	1.58 + 6	1.53 + 6
.187	2.22 + 6	1.86 + 6	1.78 + 6
.167	2.43 + 6	2.03 + 6	1.94 + 6
.150	2.50 + 6	1.94 + 6	1.82 + 6
.136	2.50 + 6	1.85 + 6	1.81 + 6
.123	2.35 + 6	1.63 + 6	1.50 + 6
.111	2.09 + 6	1.39 + 6	1.24 + 6
.101	1.70 + 6	1.08 + 6	9.25 + 5
.0912	1.28 + 6	7.31 + 5	5.80 + 5
.0831	9.10 + 5	4.72 + 5	3.15 + 5
.0755	6.49 + 5	3.39 + 5	2.47 + 5
.0688	5.67 + 5	3.62 + 5	1.53 + 5
.0627	4.08 + 5	2.93 + 5	1.59 + 5
.0572	3.00 + 5	2.39 + 5	3.89 + 4
.0523	2.03 + 5	1.68 + 5	3.37 + 4
.048	1.14 + 5	7.61 + 4	1.89 + 4
.044	8.2 + 4	5.8 + 4	-
.040	4.2 + 4	2.8 + 4	-

The relationship between Z and V (equation 2) and the fact that voltages increase in geometric ratio (equation 5) together ensure that, just as the first channel (0.840 µm) is matched with the fifth (0.483 µm) in this calculation, so subsequent channels will be similarly matched. Thus, referring to Table A-1, the value of Z for 0.207 µm (8.29 - 5) on doubling will lie between Z for .136 and .123 µm, and again closer to the former, four places down from .207 µm.

In the calculation the data is treated four channels at a time, starting from the largest size. The first round of calculation is shown in Table A-3. Columns have the following significance:

(1) particle diameter;

(2) first order concentration from Table A-2, assumed correct;

(3) percentage +2 charged particles at size 1, from Figure 3;

(4) the product of (2) and (3), is the concentration of doubly charged particles to be expected in the next four channels (.483 to .330 µm inclusive);

(5) the doubled raw count for these next four channels, taken from Table A-1;

(6) (5) - (4) the corrected raw count for these sizes;

(7) the % +1 charged particles for these sizes, again taken from Figure 3.

(8) (6) ÷ (7) is then the corrected second order concentrations for the sizes .483 to .330 µm.

TABLE A-3

Second order correction, TSI 3076, 3 l/min, first round. See text for significance of column heading numerals.

1.	2.	3.	4.
.840 µm	1.0 + 4	11.2%	1.12 + 3
.728	2.05 + 4	11.55	2.37 + 3
.633	3.87 + 4	11.8	4.57 + 3
.552	8.04 + 4	12.05	9.69 + 3

5.	6.	7.	8.	Corrected Value for
2.48 + 4	2.37 + 4	17.2%	1.38 + 5	.483 µm
5.10 + 4	4.86 + 4	18.0	2.70 + 5	.424
7.84 + 4	7.38 + 4	18.9	3.90 + 5	.373
1.22 + 5	1.12 + 5	19.8	5.66 + 5	.330

Setting up now a second table as above with .483 to .330 μm as the four entries in column (1), and their second order corrected values just found in column (2), the process is repeated in batches of four until one has stepped through all sizes and reached the second order values listed in column 3 of Table A-2, and shown in the middle curve of Figure 4.

Third Order Calculation

This begins with the observation that the mobility for .840 μm tripled (3.957 - 5) lies between mobilities for the seventh and eighth sizes down (.373 and .333 μm) and somewhat closer to the eighth (ie. seven places down). Accordingly the data is treated in batches of seven. The first round of calculation is shown in Table A-4. Contributions of +3 charged particles from diameters .840 to .373, in column (1), are attributed to the next seven diameters .330 to .167 μm.

TABLE A-4

Third order correction, TSI 3076, 3 l/min, first round. See text for significance of column heading numerals.

1.	2.	3.	4.	
.840 μm	1.0 + 4	8.0%	8.0 + 2	
.728	2.05 + 4	7.8	1.60 + 3	
.633	3.87 + 4	7.55	2.92 + 3	
.552	8.04 + 4	7.2	5.79 + 3	
.483	1.38 + 5	6.65	9.18 + 3	
.424	2.70 + 5	6.1	1.65 + 4	
.373	3.90 + 5	5.5	2.15 + 4	
5.	**6.**	**7.**	**8.**	**CORRECTED VALUE FOR**
1.12 + 5	1.12 + 5	19.8%	5.66 + 5	.330 μm
1.50 + 5	1.48 + 5	20.7	7.15 + 5	.293
1.83 + 5	1.80 + 5	21.3	8.45 + 5	.260
2.73 + 5	2.67 + 5	21.9	1.22 + 6	.232
3.53 + 5	3.43 + 5	22.4	1.53 + 6	.207
4.26 + 5	4.07 + 5	22.9	1.78 + 6	.187
4.71 + 5	4.28 + 5	23.2	1.94 + 6	.167

Column (2) lists the best available concentrations for (1), the first approximation for the four largest sizes, and for the remaining three, the second order corrected value found above. (3) is now the percentage +3 charge for the sizes in column (1) taken from Figure 3. Column (5), the raw CNC counts are the best available values, namely the corrected raw counts from the second order determination just made. The remaining columns have the same significance.

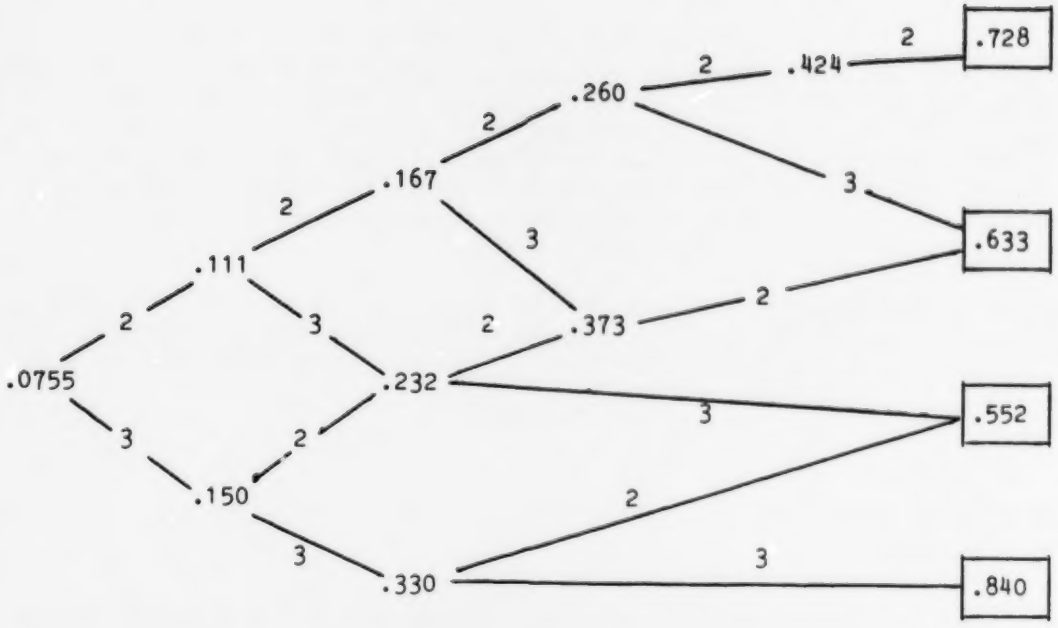
This is carried through in batches of seven. As before, the third order corrections of column (8) are carried forward to the second round and placed in column (2). Table A-2 column 4, and the lower curve of Figure 4 show the third order corrected values.

Observations

Down to a size ~0.1 μm (see Figure 4), the third order correction is hardly to be distinguished from the second, and over most of the range >0.1 μm both of these are only minor corrections to the first approximation. There are self-compensating features resulting from the skewed normal distribution observed in Figure 4. At the upper size end the % content of multiply charged particles is larger but these corrections are applied to sizes whose population is considerably greater to begin with, and make little difference. By the same token, corrections become significant in the lower size range, on the left hand side of the curve, but here the mass (though not the number) contribution to the total aerosol is small; in the applications of this instrument the mass of material penetrating a filter is the most important factor.

The method described above is open to the objection that, by applying all second order, and then all third order corrections, in sequence, certain interactions are overlooked. This was investigated, taking selected diameters and setting up a tree in which these are connected to larger diameters by second or third order effects, stepping back four or seven places respectively, until one reaches one or the other of the four largest diameters, whose concentrations are by definition assumed correct.

Such a tree is shown below. Starting from the four largest sizes (enclosed in boxes) one can apply all second and third order corrections to the raw count data for smaller sizes.



The results of this calculation are shown for a number of sizes in the last column.

TABLE A-5

SIZE, μm	1st ORDER	2nd ORDER	3rd ORDER	3rd ORDER REFINED
.330	6.15 + 5	5.66 + 5	5.66 + 5	5.62 + 5
.260	1.01 + 6	8.59 + 5	8.45 + 5	8.47 + 5
.232	1.46 + 6	1.25 + 6	1.22 + 6	1.22 + 6
.167	2.43 + 6	2.03 + 6	1.94 + 6	1.94 + 6
.150	2.50 + 6	1.94 + 6	1.82 + 6	1.85 + 6
.111	2.09 + 6	1.39 + 6	1.24 + 6	1.27 + 6
.0755	6.49 + 5	3.39 + 5	2.47 + 5	2.79 + 5

It is evident that the sequential method slightly over-estimates the 3rd order correction, but in view of the measurement precision the difference is insignificant, and the simple third order correction is entirely adequate even at sizes (.0755 μm) where this correction is large, and modifies considerably the first order approximation.

Calculation of Size Distributions

From a set of data, number concentration vs diameter, by which as always is understood aerosol concentrations in a series of diameter increments, are derived other measures of aerosol distribution, as is shown in Table A-6. Here columns A and B are diameters and third order corrected concentrations for the TSI 3076. Derivations in the succeeding columns are explained in the notes appended to the table.

TABLE A-6

Calculation of aerosol properties, TSI 3076, 3 l/min

A Size μm	B 3rd Order Conc	C $(Size)^3$ $(\mu m)^3$	D B x C - m_i	E Cumulative D - M	F A x D - $m_i d_i$	G A x B - $n_i d_i$
.840	1.0 + 4	.59270	5927	213434	4987	8400
.728	2.05 + 4	.38583	7910	207507	5758	14924
.633	3.87 + 4	.25364	9816	199597	6214	24497
.552	8.04 + 4	.16819	13522	189781	7464	44381
.483	1.38 + 5	.11268	15550	176259	7510	66654
.424	2.70 + 5	.076225	20580	160709	8727	114480
.373	3.90 + 5	.051895	20239	139859	7549	145470
.330	5.66 + 5	.035937	20340	119620	6712	186780
.293	7.15 + 5	.025154	17985	99280	5270	209495
.260	8.45 + 5	.017576	14852	81295	3861	219700
.232	1.22 + 6	.012487	15234	66443	3524	283040
.207	1.53 + 6	.008869	13570	51209	2809	316710
.187	1.78 + 6	.006539	11640	37639	2177	332860
.167	1.94 + 6	.004657	9035	25999	1509	323980
.150	1.82 + 6	.003375	6142	16964	921	237000
.136	1.81 + 6	.002515	4552	10822	619	246160
.123	1.50 + 6	.0018608	2791	6270	343	184500
.111	1.24 + 6	.001368	1696	3479	188	137640
.101	9.25 + 5	.001030	953	1783	96	93425
.0912	5.80 + 5	.000759	440	830	40	52896
.0831	3.15 + 5	.000574	181	390	15	26176
.0755	2.47 + 5	.000430	106	209	8	18648
.0688	1.53 + 5	.000326	50	103	3	10526
.0627	1.59 + 5	.000246	39	53	2	9969
.0572	3.89 + 4	.000187	7	14	-	2225
.0523	3.37 + 4	.000143	5	7	-	1702
.048	1.89 + 4	.00011	2	2	-	907

$\Sigma B = 18.4 \times 10^6$ $\Sigma D = -M$ $\Sigma m_i d_i = 76306$ $\Sigma n_i d_i = 3,349,000$

Mass median diameter Entries in Column C are cubes of the diameters listed in Column A, so that quantities derived from this column are proportional to volume and therefore to mass. Entries in Column D, the product of the corresponding lines in columns B and C, are then proportional to the mass contained in each increment of diameter. The next column lists the cumulative value of column D, beginning the addition at the bottom of the column, so that the first entry (213,434) is proportional to total aerosol mass. From this, the mass median diameter can be found, the diameter above and below which aerosol masses are equal. 213,434 ÷ 2 = 106,717 and the size corresponding to this halfway point is 0.30 μm.

Mass mean diameter is defined as $\frac{m_i d_i}{M}$. Column F is obtained by multiplying corresponding lines of A (d_i) and D (m_i). The sum of this column is 73,606, which on division by the cumulative value of E, 213,434 gives 0.36 μm as the mass mean diameter.

Similarly the number mean diameter is defined as $\frac{\Sigma n_i d_i}{N}$ in which column G, obtained by multiplying columns A and B gives values of $n_i d_i$. Its sum is divided by total number count, the sum of column B, to give 0.18 μm.

Lastly, assuming the derivation of equation 5 is correct, so that the mobility bands are contiguous, an estimate of the total mass concentration (wt/vol) of the aerosol can be made. The sum of column D is proportional to the total aerosol mass. Inserting the proper proportionality factor to convert to units of μg/l, and the density of DOP to convert volume to mass, one finds $213,434 \times \frac{\pi}{6} \times 10^{-3} \times 0.98 = 110$ μg/l.

Remarks on Significant figures The diameters of column A are carried to three significant figures, one more than justified, mainly to facilitate the juxtaposing of channels when calculations involving 2Z and 3Z, for higher order corrections, are carried out. The concentrations of column B are good at best to two figures.

The purpose in carrying the extra numbers in column D to G is to show the contrast between D-F and G. The first three of these, depending on volume, rapidly become insignificant below a diameter of 0.1 μm, so that these entries could in fact be dropped without affecting the results. Column G depends on number, not mass, and the contribution from particles below 0.1 μm is still appreciable. The derived aerosol measures are all rounded off to two figures.

It is evident that for purposes of comparison one can often use the first order approximation or even the raw data.

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